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METHODOLOGIES FOR DETERMINING THE SOURCES, CHARACTERISTICS, DISTRIBUTION, AND ABUNDANCE OF ASBESTIFORM AND NONASBESTIFORM AMPHIBOLE AND SERPENTINE IN AMBIENT AIR AND WATER

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Anthropogenic and nonanthropogenic (erosion) processes contribute to the continuing presence of asbestos and nonasbestos elongated mineral particles (EMP) of amphibole and serpentine in air and water of urban, rural, and remote environments. The anthropogenic processes include disturbance and deterioration of asbestos-containing materials, mining of amphibole- and serpentine-bearing rock, and disturbance of soils containing amphibole and serpentine. Atmospheric dispersal processes can transport EMP on a global scale. There are many methods of establishing the abundance of EMP in air and water. EMP include cleavage fragments, fibers, asbestos, and other asbestosiform minerals, and the methods employed do not critically distinguish among them. The results of most of the protocols are expressed in the common unit of fibers per square centimeter; however, seven different definitions for the term “fiber” are employed and the results are not comparable. The phase-contrast optical method used for occupational monitoring cannot identify particles being measured, and none of the methods distinguish amphibole asbestos from other EMP of amphibole. Measured ambient concentrations of airborne EMP are low, and variance may be high, even for similar environments, yielding data of questionable value for risk assessment. Calculations based on the abundance of amphibole-bearing rock and estimates of asbestos in the conterminous United States suggest that amphibole may be found in 6–10% of the land area; nonanthropogenic erosional processes might produce on the order of 400,000 tons or more of amphibole per year, and approximately 50 g asbestos/km²/yr; and the order of magnitude of the likelihood of encountering rock bearing any type of asbestos is approximately 0.0001.

Data from the United States and elsewhere pertaining to occurrence, levels of concentration, and mineral identity of amphibole and serpentine particles found in ambient environments are provided in this review. However, workplace environments are not addressed.

Amphibole and serpentine minerals are common rock-forming minerals, and as such are widespread in the rocks and soils. These can occur in a variety of growth forms, called habits, although massive, nonfibrous habits are

much more common than fibrous habits. In a small subset of the occurrences of these minerals, they are found in a special habit that possesses unique physical and chemical properties, which, before the links between asbestos and diseases such as mesothelioma, lung cancer, and asbestosis were well established, rendered them useful as building materials, and they were mined as asbestos. Asbestos may also have been incorporated into building material when it was associated with other commercially useful materials.

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In all habits, small mineral particles enter the atmosphere locally, becoming suspended as elongated mineral particles (EMP) if they attain a length to width ratio of 3:1 or greater (National Institute for Occupational Safety and Health [NIOSH], 2011). Most peer-reviewed published data concerning occurrences and concentrations of airborne and waterborne EMP were collected many years ago when asbestos use was at its height. Since the mid 1970s, mining of asbestos has virtually ceased in most places, with notable exceptions in Russia, Brazil, China, and Kazakhstan (Virta, 2012). It is likely that the ambient level of asbestos derived from use of commercial asbestos-containing materials has declined over the past 40 years despite the fact that asbestos removal activities are ongoing and continue to contribute to ambient levels. Nonasbestos EMP, however, are also found in air and water and are likely to remain in the ambient environment as population grows and land use expands; this is especially the case in arid and semiarid regions west of the Mississippi River in the United States, and as the use of sand, gravel, and crushed stone for building infrastructure continues. Important sources for ambient EMP include erosion of rock and soil, mining and use of rock containing amphibole and serpentine, anthropogenic disturbances of soil containing amphibole and serpentine, and fabrication, use, and disturbance of synthetic products containing asbestos. These sources will continue to generate EMP in the local environment, both asbestos and nonasbestos, and continue to enter the atmosphere and surface water. Although most of these particles settle out of air or water, some may be transported great distances.

BACKGROUND

Asbestos has been collectively implicated as a causal agent in malignant diseases, notably mesothelioma and lung cancer, and nonmalignant diseases including asbestosis, pleural effusions, pleural plaques, diffuse pleural thickening, rounded atelectasis, autoimmune diseases, and cardiovascular diseases (NIOSH,

2011; Broaddus et al., 2011; Bunderson-Schelvan et al., 2011; Mossman et al., 2011; Shannahan et al., 2012). It has also been inferred as a causal agent in laryngeal cancer and is suggestive for pharyngeal, stomach, and colorectal cancers (Samet et al., 2006). Epidemiological evidence and responses to asbestos in animal experiments have been recently reviewed by Lippmann (2014). Elevated occurrences of asbestos-associated disease are also reported from exposure to dusts containing other asbestosiform amphibole and serpentine minerals not mined as asbestos, including the asbestosiform varieties of the amphiboles winchite, richterite, and fluoroedenite (McDonald et al., 1986; Amandus and Wheeler, 1987; Gianfagna et al., 2003; Comba et al., 2003; Wylie and Verkouteren, 2000; Meeker et al., 2003; Sullivan, 2007) and an unusual asbestosiform habit of the serpentine mineral antigorite (Keeling et al., 2008).

These diseases are also associated with exposure to two other fibrous minerals, asbestosiform balangeroite (Gassano et al., 2005) and Turkish erionite (Baris et al., 1981). In Turkey, it appears that factors other than the properties of inhaled erionite fibers affect risk, given that the same types of exposures in towns less than 2 km apart produce vastly different disease outcomes (Below et al., 2011; Baris et al., 1981; Roushdy-Hammady et al., 2001).

The specific modes of action (MOA) by the various types of asbestos in the development of asbestos-related diseases are unknown, but both MOA and potencies differ among asbestosiform mineral types (Mossman et al., 2011; Cyphert et al., 2012; Berman and Crump, 2008, 2003; Lippmann, 2014). For example, amphibole asbestos is generally recognized as a more potent carcinogen than chrysotile (Case et al., 2011; Lippmann, 2014). It is generally agreed that not only is the toxicity of asbestos related to the physical features such as width, length, aspect ratio and effective surface area, but durability, crystallinity, surface chemistry, and reactivity are also factors that need to be considered in understanding the carcinogenicity of asbestos (Aust et al., 2011; Huang et al., 2011).

In the absence of asbestos, asbestos-related diseases are not associated with exposure to nonasbestos EMP of amphibole or serpentine in humans (Addison and McConnell, 2008; Gamble and Gibbs, 2008; Nolan et al., 1991; Ilgren, 2004; Williams et al., 2012). In addition, Mossman et al. (2011) demonstrated that cells respond differently to different habits of the same mineral, and Davis et al. (1991) noted that rodent inhalation studies using different habits of tremolite produced variable responses including no tumors when exposure contained only cleavage fragments. Intratracheal installation in rats reported by Kodavanti et al. (2014) and Cyphert et al. (2012) provides additional examples of the importance of habit. These investigators demonstrated that Libby amphibole (12% with aspect ratio [ar] < 3:1), composed of a range of habits from prismatic to asbestiform (Meeker et al., 2003), and chrysotile asbestos (2% with ar < 3:1) are more potent inducers of acute-phase proteins and produce greater lung inflammation and pulmonary injury than fragmented ferroactinolite (designated as cleavage fragments by the U.S. Geological Survey [USGS], which provided the sample) from Ontario (60% with ar < 3:1) and tremolite (undescribed habit) from El Dorado Hills, CA (66% with ar < 3:1). In addition, amphiboles and serpentine that form in habits of growth other than asbestos are not regulated under the asbestos standard by the Occupational Safety and Health Administration (OSHA, 1992).

A case was made that asbestiform fibers and nonasbestiform fragments of the same dimensions may exert similar effects in cells (Case et al., 2011). However, among natural populations, the dimensional overlap is small, occurring mainly among EMP less than a few micrometers (Wylie et al., 1985; Siegrist and Wylie, 1980; Wylie and Schweitzer, 1982) in length. These same dimensional studies demonstrated that in cleavage fragment populations, width increases with length, whereas in asbestos populations, widths are small, almost constant, and independent of length. Therefore, the hypothesis for similarity in response of cleavage fragments and asbestos fibers of

the same dimension put forward by Case et al. (2011) is unlikely to be easily evaluated. Further, until the MOA of asbestos is understood, characteristics of populations other than dimensions cannot be assumed to be unimportant.

There have been many proposals for the size and shape of durable mineral particles that have the highest probability of being carcinogenic. These results were derived from various approaches, but nonetheless are remarkably consistent: Pott (1978) proposed $l \geq 10\text{ }\mu\text{m}$, $w \leq 0.25\text{ }\mu\text{m}$; Spurney et al. (1979) proposed $l \geq 5\text{ }\mu\text{m}$, $w \leq 0.5\text{ }\mu\text{m}$; Stanton et al. (1981) found the highest correlation with tumors in rats for fibers with $l \geq 8\text{ }\mu\text{m}$, $w \leq 0.25\text{ }\mu\text{m}$; Lippmann (2014) reaffirmed his earlier proposal that mesothelioma is produced by biopersistent fibers with $l \geq 5\text{ }\mu\text{m}$, $w \leq 0.1\text{ }\mu\text{m}$, and lung cancer and asbestosis by asbestos fibers with $l \geq 20\text{ }\mu\text{m}$. Loomis et al. (2010) found $l \geq 20\text{ }\mu\text{m}$ and w between 0.25 and 1 μm were most strongly associated with lung cancer, and Berman and Crump (2003) proposed that fibers with $l \geq 10\text{ }\mu\text{m}$, $w \leq 0.4\text{ }\mu\text{m}$ were most potent for lung cancer in rats. None of these conform to the seven different definitions of fiber used to measure EMP in occupational or environmental settings (see Table 1), but all reflect that carcinogenic mineral populations contain long, thin fibers.

Some investigators maintained that short fibers cannot be dismissed as biologically inactive (Dodson et al. (2003). Others proposed that although short fibers are unlikely to induce cancer in humans, they may possibly contribute to pulmonary fibrosis (Agency for Toxic Substances and Diseases, 2003; Berman and Crump, 2003; Gibbs and Pooley, 2008; Paoletti and Bruni, 2009). This topic was reviewed by Aust et al. (2011) and Broaddus et al. (2011), who point out that lack of understanding of how asbestos leads to pleural disease indicates that all characteristics of asbestos populations need to be considered, including size, shape, mineral composition, habit of formation, and biodurability. In fact, short fibers are found in abundance among certain asbestos populations. For example, models of length

TABLE 1. Dimensional Designations for "Fiber" From Methods Commonly Employed for Analysis of Ambient Asbestos

Air monitoring methods	Fiber definitions
NIOSH 7400*	Definition 1. Optically visible particles with an aspect ratio of at least 3:1 and length greater than 5 μm
NIOSH 7402†	Definition 2. Aspect ratio of at least 3:1, length greater than 5 μm , and minimum width of 0.25 μm
Yamate‡	Definition 3. Aspect ratio of at least 3:1, no length or width restrictions
AHERA§	Definition 4. Minimum aspect ratio of 5:1 and length of at least 0.5 μm
ISO 10312** and ISO 13794††	Definition 4. Minimum aspect ratio of 5:1 and length of at least 0.5 μm . Alternate values may be employed as described in the Annex. Rules for classifying structures as bundles, clusters and matrix are also included.
ISO 14966††	Definition 5. Minimum aspect ratio of 3:1, length greater than 5 μm , and width less than or equal to 3 μm
Water monitoring methods	Fiber definition
U.S. EPA Method 100.1§§	Definition 6. Minimum aspect ratio of 3:1, largely parallel sides, and length greater than 0.5 μm
U.S. EPA Method 100.2***	Definition 7. Minimum aspect ratio of 3:1, largely parallel sides, and length greater than 10 μm

Note. Sources: *NIOSH 1989.

†NIOSH 1994.

‡Yamate et al. 1984.

§U.S. EPA 2002.

**ISO 1995.

††ISO 1999.

‡‡ISO 2002.

§§Chatfield and Dillon 1983.

***Brackett et al. 1994.

distributions of crocidolite from Cape Province, South Africa, and from the Hammersley Range of Australia predict that for every fiber of crocidolite longer than 5 μm there are about 10^3 fibers less than 5 μm (Figure 1 in Wylie, 1993). All occupational exposures on which risk assessments have been based include fibers longer than 5 μm , as well as short fibers, so the issue of which part of the population is most biologically active will likely remain unresolved for some time. The work of Berman and Crump (2008), which evaluated the effects of dimension and mineralogy across study environments, provides a promising approach to address this problem.

Many investigators advocated for a consensus on the dimensions of EMP that are most strongly associated with asbestos-related diseases and that may be used as markers for exposure (Institute of Medicine [IOM], 2009; Case et al., 2011) so that airborne concentrations can be used for assessing risk. If markers of exposure were established, it should not be assumed that these and only these particles are biologically active. Rather, their presence indicates an airborne population of potentially carcinogenic fibers. The argument persists that one cannot abandon the current regulatory criteria

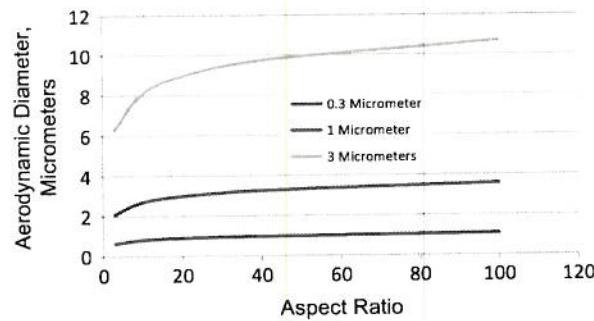


FIGURE 1. Relationship between aspect ratio and aerodynamic diameter for particles with widths of 0.3, 1.0, and 3.0 μm , calculated from the equation of Burke and Esman (1978) by John Richards (personal communication).

and methodology for measuring exposure levels (optical microscopy) because occupational risks were based on them, and using TEM for exposure assessment is impractical (Lippmann, 2014). However, regulatory criteria for airborne fibers remain nonspecific for asbestos and are therefore problematic for assessing risk outside an occupational setting. Further, TEM is frequently used to measure environmental exposure (see Tables 3–11). Building consensus on such analytical targets and TEM methodology likely requires additional studies.

Extrapolating risk established from occupational exposure to low level exposures is difficult and fraught with uncertainty (Case et al., 2011). Controlling for exposure level, exposure duration and cohort age, White et al. (2008) demonstrated that environmental exposure to crocidolite resulted in excess mesothelioma in South Africa but environmental exposure to chrysotile did not, emphasizing the point that the identification of the minerals in the exposure is an important element in risk assessment. Bourdes et al. (2000) employed a meta-analysis to distinguish the relative risks from (1) high “neighborhood” exposures, which resulted from asbestos mining and manufacturing close to the place of residence, (2) household contact with asbestos workers, and (3) exposures from the use of asbestos-containing soils as whitewash. Although risk could be estimated from these high environmental exposures, a dose-response model was not proposed, and Bourdes et al. (2000) concluded that available data are insufficient to estimate excess risk from exposures encountered by the general population. Data indicated that (1) exposures are from different sources of environmental asbestos (natural vs. mining), (2) there are no epidemiological studies on more common low-level exposures, (3) there are publication biases such as the fact that negative studies are rarely published, (4) occupational exposure may have occurred, and (5) there is the problem of misdiagnosis. Bourdes et al. (2000) did not consider the problem of particle identification in environmental exposure assessment, which this review addresses.

DEFINITIONS

Asbestos, Asbestiform, Fibers, and EMP

The term “asbestos” as used in this review is defined as any amphibole in the asbestiform habit with a chemical composition that places it in one of four chemical series as defined by Leake et al. (1997): tremolite–ferroactinolite, cummingtonite–grunerite, anthophyllite or riebeckite–magnesioriebeckite. The term

TABLE 2. Asbestos Names

Mineral type	Regulatory name	Mineral name
Serpentine	Chrysotile asbestos	Chrysotile
Amphibole	Tremolite asbestos	Tremolite
	Actinolite asbestos	Actinolite
	Anthophyllite asbestos	Anthophyllite
	Crocidolite asbestos	Riebeckite
	Amosite asbestos	Cummingtonite-grunerite

“asbestos” is also applied to the serpentine-group mineral chrysotile. A full discussion of the mineralogy of asbestos is provided by Zoltai (1981) and Veblen and Wylie (1993). To avoid confusion, in this review the term “asbestos” is restricted to these minerals, reflecting its origins as a commercial term and its designation as such in regulatory policy. Table 2 presents the names of the six minerals that are specifically named in regulations. Other amphibole compositions may also occur in an asbestiform habit and rarely, also the serpentine mineral antigorite. Such occurrences are referred to simply as asbestiform.

The term “asbestiform” applies to a habit of mineral growth in which flexible fibrils of fine width (<0.5 µm) form in bundles of parallel fibrils. Within the bundles, one crystallographic axis of the fibrils is parallel (parallel to length), but in the other crystallographic directions, the fibrils are not aligned and are readily separated (Alario Franco et al., 1977; Heinrich, 1965; Zoltai, 1977; Cressey et al., 1982). The high aspect ratio of these fine fibrils and fibril bundles enhances their flexibility and tensile strength (Hodgson, 1965; Zoltai, 1981). Minerals other than amphibole and chrysotile may occur in an asbestiform habit, such as talc, brucite, balangeroite, erionite, carlosturanite, and antigorite.

With the light microscope, the asbestiform habit in a population of fibers observed in bulk samples is generally recognized by the following characteristics (U.S. Environmental Protection Agency [EPA], 1993):

- Mean aspect ratios (length/width) ranging from 20:1 to 100:1 or higher for fibers longer

than 5 μm . Aspect ratios needs to be determined for individual fibers (called fibrils), not for bundles of fibers.

- Very thin fibrils, usually less than 0.5 μm in width.
- Two or more of the following:
 - Parallel fibers occurring in bundles.
 - Fiber bundles displaying splayed ends.
 - Matted masses of individual fibers.
 - Fibers showing curvature.

The U.S. EPA further states that although occasional particles can have aspect ratios of 10:1 or less, it is unlikely that the asbestos component of bulk materials would be dominated by particles (individual fibers) having aspect ratios of <20:1 for fibers longer than 5 μm .

In a hand specimen of asbestos, the asbestiform habit is clearly evident in the long, thin, flexible single fibrils or bundles of fibrils that are easily separated by shear stress applied by hand. The asbestiform habit persists in powdered samples, and bundles of very thin fibrils are normally visible under the optical microscope as described earlier. Although individual fibrils may be well below the resolution of the optical microscope, in bundles they produce a set of optical properties that are characteristic of the habit (Heinrich, 1965; Wylie, 1979; Verkouteren and Wylie, 2002). When subjected to mechanical forces sufficient to generate airborne particles, bundles are separated into smaller bundles or individual fibrils. Fibrils, both single and in small bundles, less than 5 μm in length and less than 0.5 μm in width predominate in air, water, and lung tissue populations; fibers longer than 100 μm are uncommon (Timbrell et al., 1970; Friedrich et al., 1992; Dufresne et al., 1996; Dodson et al., 1990; Gibbs et al., 1990; Gibbs and Hwang, 1980; Addison and McConnell, 2008; Wylie et al., 1993; Pooley and Clark, 1980; Warnock, 1989; Lippmann and Timbrell, 1990).

The minerals listed in Table 2 are considered to be rock-forming minerals. This indicates that they are among a 100 or so minerals (out of many 1000s) whose occurrence is widespread in rock that constitute the earth's crust. Minerals in rocks are normally not

asbestiform. In fact, the occurrence of minerals in the asbestiform habit is generally uncommon (Klein, 1993). When crushed or otherwise broken, nonasbestiform amphibole breaks preferentially along certain parallel planes of weakness, resulting in fragmented particles that are elongated. These are referred to as amphibole *cleavage fragments*. Serpentine rock may also form elongated cleavage fragments when crushed or broken, but less commonly than amphibole.

Fibers reported in air monitoring studies should not be confused with the mineralogical term *fiber*, which means "resembling an organic fiber." In a mineralogical sense, fibers attain their shape when the mineral nucleates and grows. Although it is true that all asbestos is composed of fibers, it is not the case that all fibers are asbestos, as many minerals, under certain conditions, may form fibers.

In some environments, amphiboles and serpentine minerals may form as finely acicular fibers that are not asbestiform (Dorling and Zussman, 1987). Some may carry special designations. For example, serpentine might form a variety known as *picrolite*, and acicular fibers of amphibole are referred to as *byssolite* (Dorling and Zussman, 1987). Fibrous habits are uncommon and amphibole are normally found in prismatic or massive habits and serpentine is normally massive.

Habits other than asbestiform result in brittle behavior of amphibole and serpentine. That is, when crushed, they may form cleavage fragments, some of which will meet one of the definitions for fiber used in air monitoring (Table 1). From a mineralogical perspective, however, the act of crushing nonasbestiform minerals cannot create asbestos fibers.

Is It an Asbestos Fiber or Is It a Cleavage Fragment?

In 2011, the National Institute of Occupational Safety and Health (NIOSH) formally recognized that approved methods of occupational monitoring for asbestos EMP do not distinguish asbestos from other habits of amphibole and serpentine (NIOSH, 2011).

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In light of this reality, the term *EMP* was suggested by NIOSH (2011) to be an inclusive term that applies to all elongated mineral particles with a minimum aspect ratio of 3:1. *EMP* may be asbestos fibers, asbestiform fibers, other nonasbestiform mineral fibers, or cleavage fragments. Analytical protocols apply a variety of dimensional criteria for monitoring exposure to *EMP* (seven are provided in the following), but all report data as fibers per cubic centimeter (f/cm^3) for regulatory purposes or risk assessment. In this review, where data on airborne or water concentrations of *EMP* are given in units of fibers per cubic centimeter, the protocol for counting a particle is indicated where available and the particles are referred to as *EMP*. Where the source of the *EMP* is known or suspected, it is designated by mineral name and habit or habits if known. Asbestiform and other habits of the same mineral may occur together in almost any proportion, complicating identification.

In an occupational setting where disturbance of commercial asbestos-containing building materials results in the generation of dusts, airborne amphibole or serpentine *EMP* are highly likely to be asbestos. Other *EMP*, such as cellulose, gypsum, or fiberglass, may also be present, and some cleavage fragments of amphibole or serpentine may have accompanied the asbestos. However, in many other environments, cleavage fragments might predominate. Amphibole cleavage fragments found in air and water samples have straight parallel sides, but also on average lower aspect ratios and much greater width than asbestos *EMP* of the same length (Siegrist and Wylie, 1980; Wylie and Schweitzer, 1982; Wylie et al., 1985; Harper et al., 2012). They may also exhibit stepped terminations. Elongated serpentine fragments tend to be less regularly prismatic, although they can meet criteria for *EMP* (Wylie and Bailey, 1992). The differences in dimensional characteristics of asbestos relative to cleavage fragments have been used to discriminate these populations (Siegrist and Wylie, 1980; Virta et al., 1983; Wylie et al., 1985; Van Orden et al., 2008; Chatfield, 2013; Harper et al., 2012). These methods were developed

from studies of populations that contain a large number of particles with lengths that extend over several orders of magnitude, and the problem of consistently identifying a single particle as a fiber or fragment is unresolved. Mixed populations present particular issues, although the fact that width is independent of length in asbestos populations might be used to identify a fiber subpopulation if sufficient numbers of fibers are present.

It is unfortunate that cleavage fragment *EMP* have been labeled as fibers and equated with asbestos based solely on arbitrary dimensional criteria. Such a practice adds uncertainty to exposure data that might be used to help understand risk from inhalation of mineral particles of all types. Given the low concentrations of particles in most ambient environments, particles that meet various fiber definition in use should be referred to only as *EMP* unless they can be established as coming from a known source of asbestos or cleavage fragment or can be identified as chrysotile by transmission electron microscopy (TEM) analysis (discussed further).

MEASUREMENT OF AIRBORNE AND WATERBORNE EMP**Air**

Various overlapping and competing methods are available to assess the concentrations of airborne *EMP*. Data they provide are shaped by the capabilities and limitations of two analytical techniques used—optical microscopy and electron microscopy—and by differing definitions of what is and is not designated as a fiber, making comparability between data sets difficult.

All analytical protocols provide an index of total concentration because they all exclude some part of the population by imposing limitations of length, width, and/or aspect ratio on particles counted as fibers. These indices differ, and for the same absolute concentrations, their magnitudes may vary by orders of magnitude, although they are all usually expressed

in the same unit of measure, fibers per cubic centimeter (f/cm^3).

The most common methods used for ambient particle analysis, the fiber definitions they use, their analytical limitations, and their most common uses are described next. In effect, the multiple methods described below use five different definitions for assessing concentrations of airborne fiber. Investigators may alter the counting rules in order to evaluate the number of particles meeting several fiber definitions.

NIOSH 7400 (NIOSH, 1989) and NIOSH 7402 (NIOSH, 1994) NIOSH 7400 uses phase-contrast optical microscopy (PCM). Visible EMP with an aspect ratio of 3:1 and length $>5\ \mu m$ are designated *fibers* (definition 1) and concentrations are expressed as f/cm^3 (note the difference between this designation of fiber and the mineralogical definition given earlier). There is no stated width limitation, but because only particles with widths greater than about $0.2\ \mu m$ are visible by light microscopy (this width varies somewhat with index of refraction and mineral type), there is an effective lower limit of width. Because of the abundance of asbestos fibers with widths less than $0.2\ \mu m$, especially in chrysotile and crocidolite populations (Wylie et al., 1993), asbestos may be overlooked. Mineral identity cannot be established by PCM; nonetheless, EMP are normally reported as asbestos fibers. This is the primary method for assessing occupational exposure where asbestos is being used or removed, but it is essentially useless for analysis of ambient air and water.

NIOSH 7402 is a supplemental method of NIOSH 7400. Transmission electron microscopy (TEM) is used to count EMP meeting the same length and aspect ratio criteria as NIOSH 7400. However, the *fiber* designation includes a minimum width of $0.25\ \mu m$ (definition 2). The fiber concentration is expressed as f/cm^3 and termed PCM equivalent (PCME) because $0.25\ \mu m$ approaches the lower limit of resolution following NIOSH 7400 procedures. The comparisons are not exact because visibility and resolution is often not the same (Wylie, 1985). The analytical TEM provide data

on chemical composition and atomic structure that can be used to distinguish amphibole and serpentine EMP from other minerals. In order to eliminate ambiguities in mineral identification based on chemical composition determined by using energy-dispersive x-ray analysis (EDXA, also EDS) this and other TEM methods require the use of standards. Further, many silicates display similar electron diffraction patterns that can only be differentiated by careful pattern indexing; simple pattern characteristics such as a $5.2\text{-}\AA$ spacing of layer lines are insufficient because this is a characteristic common in most silicate EMP and not specific for amphibole. These cautions apply to all TEM methods.

Both NIOSH 7400 and NIOSH 7402 are effective in counting cleavage fragments of amphibole and serpentine because airborne cleavage fragments of these minerals that are longer than $5\ \mu m$ are normally wider than $0.2\ \mu m$ (Wylie et al., 1993). However, both methods exclude many asbestos fibers, a large proportion of which have widths less than $0.25\ \mu m$ in width (Wylie et al., 1993).

Yamate et al. (1984) method Yamate et al. (1984) proposed a counting protocol for an analytical TEM. All EMP having an aspect ratio greater than 3:1 are designated as *fibers* (definition 3) and concentrations are expressed as f/cm^3 . There are no limits on length or width, so the protocol produces higher levels of concentration for the same absolute concentration than NIOSH 7400 or 7402. The lack of a length criterion leads to lab-to-lab inconsistencies in particle counting (Chatfield, 2000; Steel and Small, 1985) and concentration data, and the method does not provide an adequate assessment of the abundance of longer fibers, potentially the most biologically active.

The Asbestos Hazard Emergency Response Act (AHERA) method This uses an analytical TEM protocol that addresses the inconsistencies of EMP counting in the Yamate method (U.S. EPA, 2002). Only EMP with a minimum aspect ratio of 5:1 and equal to, or longer than $0.5\ \mu m$, are designated as *fibers* (definition 4) and concentrations are expressed as f/cm^3 . The use of the analytical TEM enables amphibole and serpentine EMP to be distinguished from other

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mineral EMP, but the fiber designation does not exclude EMP cleavage fragments. The AHERA method was designed to assess cleanliness of buildings (especially school buildings) following asbestos abatement and is not generally applicable to other environments. The inclusion of short fibers of unlikely carcinogenic potential, as is the case for the Yamate et al (1984) method, is problematic for risk assessment.

International Standards Organization (ISO) method 10312 The International Standards Organization (ISO) method 10312 (International Standards Organization, 1995) is an analytical TEM-based method intended for use in environments with asbestos, elongated mineral particles, and organic fiber. The protocol designates an EMP with a 5:1 aspect ratio and a minimum length of 0.5 μm as a *fiber* (same as AHERA). (There is an annex to this method in which aspect ratio is adjusted to 3:1.) Data are reported as structures/ cm^3 (s/cm^3), although they may be referred to as fibers in the literature. Alternate fiber definitions are sometimes employed to derive PCM equivalent (PCME) data. ISO 10312 is one of the most common methods used to compile ambient asbestos PCME concentration data. Despite the higher aspect ratio and lower lengths limits, cleavage fragment EMP may be included and ISO states the method cannot distinguish fibers from other EMP.

International Standards Organization (ISO) method 14966 International Standards Organization (ISO) method 14966 (ISO, 2002) is a scanning electron microscopy (SEM) method used for fibers in air. Fibers are defined as particles longer than 5 that have an aspect ratio of at least 3:1 and a width less than or equal to 3 μm (*definition 5*). Mineral identification is based on qualitative EDS. The method does not distinguish asbestos fibers from cleavage fragments.

ISO 13794 Sometimes one of the preceding methods (in particular the Yamate method or ISO 10312) uses an indirect transfer sample preparation technique that can change fiber size distribution and increases fiber number (Lee et al., 1995; Sahle and Laszlo, 1996). ISO 13794 (ISO, 1999) is the formal name for this

method. Data from the indirect approach have been expressed as nanograms per cubic meter (ng/m^3) as well as f/cm^3 or structures/ cm^3 . Structures may include fibers, bundles, matrices, or clusters. Concentrations expressed as ng/m^3 are not directly comparable to concentrations expressed as f/cm^3 . In the accompanying tables, the technique is referred to as *indirect TEM*.

Water

There are two standard methods for determining asbestos in water, one for all particles longer than 0.5 μm and the other for EMP longer than 10 μm . These are the U.S. EPA method 100.1 (Chatfield and Dillon, 1983) and U.S. EPA method 100.2 (Brackett et al., 1994), respectively. Both use an analytical TEM and designate all particles of amphibole or serpentine that have an aspect ratio of 3:1 or greater and largely parallel sides as fibers. In method 100.1, EMP of greater than 0.5 μm in length are included as *fibers* (*definition 6*), whereas U.S. EPA method 100.2 includes only EMP longer than 10 μm (*definition 7*). The U.S. EPA drinking water standard is 7 millions of fibers per liter (MF/L) longer than 10 μm (U.S. EPA, 2014a), although at the time of this writing the U.S. EPA information page on asbestos in drinking water is ambiguous about the length criterion ([http://water.epa.gov/drink/contaminants/basicinformation/asbestos.cfm](http://water.epa.gov/drink contaminants/basicinformation/asbestos.cfm)).

In U.S. EPA methods, particles in a representative subset of EMP are identified as amphibole or serpentine based on selected area electron diffraction (SAED). Once established as an amphibole, the mineral name rests primarily on its chemical composition. The reliability of a designation of a particle as serpentine or chrysotile is also improved if compositional information is available. As stated earlier for the air methods, TEM equipped with energy dispersive x-ray analysis (EDS) provides a qualitative chemical analysis, although the elimination of ambiguities requires consideration of uncertainties and the use of standards, and mineral identification based on SAED requires more than simple pattern characteristics such as layer

line spacing. Like all air methods, the criteria used to designate a particle as a "fiber" applies to both cleavage fragments and asbestos fibers of amphibole. The distinctive characteristics of chrysotile observed by TEM normally enable it to be readily distinguished from other forms of serpentine. Data are reported in terms of millions of fibers per liter (MF/L) or sometimes f/cm³.

Lung Tissue

There is no standard method of analysis employed in the study of the mineral content of lung tissue. Tissue must be treated to liberate fibers for analysis and a number of methods have been employed. In addition, there may be differences in concentration data that depend on sampling location in the lung, the type of microscope employed, and the choice of fiber definition used (Churg, 1993). The small size of the fibers means that TEM is normally used for analysis. Analytical TEM helps to determine mineral identity through SAED and EDS. As is the case in air and water analysis, the dimensional criteria for designation as fiber vary among studies. Lung tissue studies demonstrated the presence or absence of airborne respirable EMP, and, although the concentration of fibers per gram lung tissue is some indication of exposure, it cannot be used as part of a quantitative assessment. Gibbs and Pooley (2008) summarized EMP content from a number of lung studies covering control subjects (no known asbestos exposure) and found averages of 0.007 to 11.2 "fibers" per gram dry lung. In general, data indicated that chrysotile and amphibole EMP are higher in urban than rural areas, but that lung tissue in general contains a greater number of nonasbestos EMP in a ratio of 4:1 or 5:1 over asbestos.

Limitations in the Analysis of Ambient Asbestos

The large majority of fiber concentration data that have been published are based on one of the methods described in the preceding. Fiber concentrations are not comparable,

even when the same unit of measure is used to report concentrations (e.g., f/cm³), because of seven different definitions for the term "fiber."

Further, the identification of particles remains an issue of concern in all methods. First, dimensional definitions alone cannot distinguish between asbestos and cleavage fragments on a particle-by-particle basis because the definition of asbestosiform is based on the properties of populations of mineral particles. Second, there is no technique that can be used with phase-contrast optical microscopy by which amphibole and serpentine can be distinguished from each other, or from other minerals, and the method cannot determine which type of amphibole or serpentine mineral is present. Third, chemical analyses and electron diffraction patterns produced by TEM analysis are only reliable indicators of amphibole minerals if ambiguities are eliminated by the use of standards for EDS and careful indexing of SAED patterns. Unfortunately, the details of TEM analytical protocols are frequently omitted in published studies. These issues provide uncertainties of such magnitude that this review refers to all "fiber" concentrations as EMP concentrations. When a TEM or other form of analytical method has been used that has the capacity to identify particular minerals, the mineral identification is accepted as likely, with the understanding that this designation rests on the uncertainties just described.

Whatever definition of "fiber" is used in assessing concentrations of EMP, the methods provide only an index of concentration. Further, using the same definition, the proportion of the population included in the concentration estimate differs among asbestos types as well as between asbestos and the nonasbestiform EMP of the same minerals. For example, TEM studies showed that an index of exposure of airborne particles longer than 5 µm with widths of 0.25 µm or greater includes as little as 10% of chrysotile, 6% of crocidolite, and 30% of amosite EMP present in the sample (Wylie, 1984). On the other hand, the same index of exposure includes 100% of 5-µm-long amphibole cleavage fragment EMP (Wylie et al., 1985).

Ambient data are also impacted by low concentrations of EMP. A selection of available data for ambient air concentrations from a variety of sources in a variety of environments is presented in Tables 3–10. Although there is no absolute lower detection limit for EMP in air or water, the ability to identify and count EMP accurately and obtain reliable data is a function of several factors, including (1) sample volumes sampled on a single filter, (2) effort expended by the analyst in examining the sample, (3) concentrations and characteristics of other suspended particulates, (4) meteorological and other conditions that impact the production of dust or entrainment of waterborne particles, and (5) the analytical method(s) and protocols employed. All of these factors vary from study to study.

Even within a given study, the variance in data can be quite large. Lee and van Orden (2008) reported concentration data from thousands of measurements taken inside and outside buildings that were known to contain asbestos. Data were obtained by a single lab from a detailed analysis of air-monitoring filters following the same TEM protocol, and using EDS and SAED for mineral identification. Data included amphibole and chrysotile particles with length to width ratios of at least 3:1. The concentrations are expressed according to specific dimensional characteristics that reflect the variability in definitions as described above. Some of these data are presented in Table 3. The differing definitions produce estimates of the same concentration of airborne

EMP that may vary over several orders of magnitude. They also demonstrate that there is a high variance in such data, whatever the definition.

SOURCES AND MEASURED CONCENTRATIONS OF AMPHIBOLE AND SERPENTINE IN AIR

Erosion of Naturally Occurring Asbestos (NOA), Amphibole, and Serpentine

Normal process of weathering and erosion contribute large numbers of particles to the atmosphere. Klein (1993) estimated that more than 2×10^9 tons of particulate is contributed annually to the global aerosol. Away from urban areas and other sources of anthropogenic dust, 1 m³ of air contains approximately 100,000 particles that are between 1 and 10 µm, and 300 million particles that are between 0.1 and 0.01 µm. These particles are ultimately derived from the rock cropping out on Earth's surface and are composed of a variety of minerals.

All asbestos forms by geological processes. However, when it occurs in its original location in rock, it is referred to as naturally occurring asbestos, or NOA. NOA may be found at or near former asbestos mine sites and prospects, sometimes at other mining locations, and in rock outcrop unrelated to mining. Comprehensive reports provide detailed literature reviews of asbestos mines and other reported occurrences of asbestos in 39 states

TABLE 3. Different Expressions of the Same Concentration of Airborne Asbestos Fiber (Data From Lee and Van Orden, 2008)

Number of samples measured	All buildings		Public and commercial		Outdoor	
	3979		1336		1678	
	Median	90th Percentile	Median	90th Percentile	Median	90th Percentile
I ≥ 5 µm, w ≥ 0.25 µm (PCME) (f/cm ³)	BD ^a	0.00018	BD ^a	BD ^a	BD ^a	BD ^a
I ≥ 5 µm, ar ^b ≥ 3, all widths (f/cm ³)	BD ^a	0.00054	BD ^a	0.00033	BD ^a	BD ^a
ar ≥ 3 (Yamate) (f/cm ³)	0.00149	0.02383	0.00059	0.00598	BD ^a	0.00324
I ≥ 0.5 µm, ar ≥ 5 (AHERA) (f/cm ³)	0.00105	0.01298	0.00053	0.00328	BD ^a	0.00294
TEM indirect (ng/m ³)	0.032	3.552	0.006	1.721	BD ^a	0.031

^aIn 50% or more samples, no fibers meeting the specified definitions were detected.

^bar = length/width.

TABLE 4. Representative Concentrations of Nonoccupational Exposures to Amphibole and Serpentine EMPs derived from Crushing Rock

Reference	Location	Protocol	Concentration	Likely source	Probable EMP identity
A. Source: Mining Excavation and Use of Rock Not Known to Contain Asbestos					
Wilson et al. (2008)	Silver Bay, Minnesota 1975	$l > 5 \mu\text{m}$; ar $\geq 3:1$ TEM	0.0048 f/cm ³	Iron ore milling	Grunerite and actinolite fragments
Wilson et al. (2008)	Silver Bay, Minnesota 1998	$l > 5 \mu\text{m}$; ar $\geq 3:1$ TEM	0.00014 f/cm ³	Iron ore milling	Grunerite and actinolite fragments
B. Source: Mining, Excavation and Use of Rock Known to Contain Asbestos					
Sebastien et al. (1986)	Quebec mining towns	NIOSH 7402 PCME	0.007–0.026 f/cm ³	Chrysotile mines	Chrysotile and minor tremolite
Anastasiadou and Gidarakos (2007)	Greece, residential	NIOSH 7400 PCM	0.08–0.18 f/cm ³	Asbestos mines	Asbestos and cleavage fragment
Luo et al. (2003)	Da-you China residential	PCM	6.6–254 f/cm ³	Riebeckite-bearing rock and mud	Riebeckite fibers and fragments
Luo et al. (2003)	Da-you China, residential	ISO 14966	1.8–413 mg/m ³	Riebeckite-bearing rock and mud	Riebeckite fibers and fragments
Cattaneo et al. (2012)	Valmalenco Italy ambient	SEM w $< 0.25 \mu\text{m}$ indirect	0.0053 f/cm ³	Serpentinite quarry	Chrysotile
Rohl et al. (1977)	Montgomery County, MD	NIOSH 7400 PCM	0.002–3 f/cm ³	Crushed serpentinite	Serpentine
Sakai et al. (2001)	Japan	ar = 3:1, l $> 0.2 \mu\text{m}$	0.384–0.447 f/cm ³	Serpentinite quarries and outcrops	Chrysotile, tremolite, and 30% other
Januch and McDermott (2004)	Libby, MT	NIOSH 7402 PCME	0–0.045 f/cm ³	Vermiculite mining	Amphibole asbestos and cleavage fragments
Price (2008)	Libby residents average	PCM	0.004–1.622 f/cm ³	Vermiculite mining waste	Amphibole asbestos and cleavage fragments
Adgate et al. (2011)	Libby residents active	Model values	0.002–1.720 f/cm ³	Vermiculite mining waste	Amphibole asbestos and cleavage fragments
Spurney et al. (1980)	Germany	Unknown	0.00001–0.0001	Crushed stone quarry	Actinolite

TABLE 5. Representative Concentration of Airborne EMPs Following Soil Disturbance

Reference	Location	Protocol	Concentration	Likely source	Probable EMP identity
Ladd (2005) normal use	El Dorado Hills, CA	PCME	0.00099–0.0101 s/cm ³	Disturbed soils containing amphibole	Amphibole
Ladd (2005) normal use	El Dorado Hills, CA	0.5 µm ar ≥ 3	0.00098–0.020 s/cm ³	Disturbed soils containing amphibole	Amphibole
Ladd (2005) aggressive use	El Dorado Hills, CA	PCME	0.00099–0.110 s/cm ³	Soils containing amphibole	Amphibole
Ladd (2005) aggressive use	El Dorado Hills, CA	0.5 µm ar ≥ 3	0.00675–1.09 s/cm ³	Soils containing amphibole	Amphibole
U.S. EPA (2008)	ISO 10312, l > 5 µm	ISO 10312, l > 5 µm	0.005–0.44 f/cm ³	Disturbed soils containing chrysotile	Chrysotile
Metintas et al. (2002)	NIOSH 7400 PCM	NIOSH 7400 PCM	0.009 to 0.040 f/cm ³	Soils used for whitewash	Tremolite
Cooper et al. (1979)	NIOSH 7400 PCM	NIOSH 7400 PCM	0.3–5.6 f/cm ³	Soils over serpentinite bedrock	Primarily chrysotile
Nassaro et al. (2012)	Unspecified	Unspecified	1 to >200 f/cm ³	Soils over serpentinite bedrock	Chrysotile
Constantopoulos (2008)	Unspecified	Unspecified	>200 f/cm ³	Preparation of whitewash	Tremolite
Constantopoulos (2008)	Mesovo, Greece	Unspecified	1–4 f/cm ³	Cleaning in whitewashed rooms	Tremolite
ATSDR (2009b)	Sapphire Mine, NC	ISO 10312 PCME	0.0065–0.036 f/cm ³	Shoveling soils	Anthophyllite

TABLE 6. Representative Concentrations of Airborne EMPs From Manufacture, Use, and Damage of Asbestos-Containing Materials (ACM)

Reference	Location	Protocol	Concentration	Likely source	Probable EMP identity
Awad (2011)	Egypt	PCM	0.002–5.4 f/cm ³	Concrete manufacturing plants	Asbestos, other
Maule et al. (2007)	Casals, Italy	PCME	0.001–0.011 f/cm ³	Concrete manufacturing facility	Asbestos, other
ATSDR (2009a)	Illinois	ISO 10312	bd (<0.0005)–0.0010 f/cm ³	Asbestos-containing waste	Asbestos, other
van Orden et al. (1995)	Loma Prieta CA (earthquake)	NIOSH 7402 PCME	0.0001 f/cm ³	In- and outside damaged buildings	Asbestos, other
Webber et al. (1988)	Woodstock NY residential	Yamate	0.05–2.8 f/cm ³	Detonating AC water pipes	94% Chrysotile, 6% amosite

in the conterminous United States (Van Gosen, 2005, 2006, 2007, 2008, 2010; Van Gosen and Clinkenbeard, 2011), and provide the number of known NOA deposits as 860. East of the Mississippi River, locations follow the Appalachian Mountain Chain. West of the Mississippi they are widely distributed and occur in every state except Nebraska and North Dakota. Outside of the southwestern states and California, where a large number of chrysotile locations are known, 65% of reported occurrences are asbestosiform amphibole. These deposits range from small vein occurrences to larger accumulations. Given that some asbestos deposits, both large and small, likely remain undiscovered or unreported, Van Gosen's (2005, 2006, 2007, 2008, 2010) reports provide a minimum estimate of the occurrences.

Amphibole asbestos is almost always associated with more widespread, massive amphibole. Chrysotile is almost always associated with massive serpentine minerals. Certain types of rock normally contain amphibole or serpentine, (e.g., mafic and ultramafic rock* of all types, marble, and iron-rich quartzite), but amphibole is also a common mineral in granite (*sensu lato*) and granitic gneiss. Maps of potential asbestos occurrences, and abundance of amphibole- and serpentine-bearing rock in general, show that the likelihood of finding amphibole and/or serpentine at some level is high over large portions of the continental United States, most particularly in the western

United States, the Appalachian Mountains, and the ancient cratons underlying Minnesota, Michigan, and New York (Lee et al., 2008).

The abundance of amphibole on the earth's surface can be estimated in a number of ways. First, one needs to consider interpretation of soil analyses. Thompson et al. (2011) noted that soils from 49 states of the United States contain amphibole. These data are based on determinations of the mineral content of individual soil horizons based on a point count of 300 individual particles. Such a widespread occurrence of amphibole does not closely correlate with the occurrences of igneous and metamorphic rocks known to contain amphibole. (See map of United States showing amphibole-containing pedons, in Figure 1 of Thompson et al. [2001].) They also report that in 41 states, 10% or more of the soils contain amphiboles, and, overall, 13% of soil pedons contain amphibole. Data suggest that 10% or more of the surface area of the conterminous United States may be a source for airborne and waterborne EMP of amphibole or serpentine.

Another approach to estimating the abundance of amphibole is based on studies of Peucker-Ehrenbrink and Miller (2002), who quantitatively analyzed the areal distribution of bedrock for the conterminous United States based on data from the corresponding U.S. Geological Service (USGS) digitized geologic map. They found that sedimentary rocks underlie 83% of this region, with the total land area given as 7,782,952 km². Igneous and metamorphic lithologies occupy 17% of this land area. Blatt and Jones (1975) estimated, based

*Mafic and ultramafic refer to rocks rich in iron and magnesium.

TABLE 7. Representative Concentrations of EMPs in Indoor Air in Buildings with Asbestos-containing Materials (ACM)

Reference	Location	Protocol	Concentration	Type of ACM	Probable EMP identity
Burdette and Jaffrey (1986)	UK 43 buildings 1983–1985	$l > 5 \mu\text{m}$; $ar > 3:1$; $w < 3 \mu\text{m}$ TEM	< 0.0001–0.002 f/cm ³	Chrysotile with minor amphibole	
Burdette and Jaffrey (1986)	UK 43 buildings 1983–1985	$l > 5 \mu\text{m}$; $ar > 3:1$; $w < 3 \mu\text{m}$ PCM	<0.001–0.017 f/cm ³	EMPs of any material	
Burdette and Jaffrey (1986)	UK 43 buildings 1983–1985	$ar > 3:1$; $w < 3 \mu\text{m}$ TEM	< 0.001–0.25 f/cm ³	Chrysotile with minor amphibole	
CPSC (1987)	San Francisco, CA	ISO 10312	0.005 s/cm ³	Cementitious	
CPSC (1987)	San Francisco, CA	ISO 10312, $l > 5 \mu\text{m}$	BD	Cementitious	
CPSC (1987)	Cleveland, OH	ISO 10312	0.0055 s/cm ³	Insulation and pipe covering	
CPSC (1987)	Philadelphia, PA	ISO 10312	0.005 s/cm ³		
U.S. EPA (1986)	U.S. federal buildings	NIOSH 7402, $l > 5 \mu\text{m}$	0.00028–0.00056 f/cm ³		
Gazzi and Crockford (1987)	United Kingdom	ISO 10312, $l > 5 \mu\text{m}$	0.0001–0.0025 f/cm ³	Amosite board	
Guillemin et al. (1989)	School buildings	Indirect TEM, $l > 5 \mu\text{m}$	0.00012–0.00859 s/cm ³		
Guillemin et al. (1989)	Nonschool buildings	Indirect TEM, $l > 5 \mu\text{m}$	0–0.0048 s/cm ³		
Guillemin et al. (1989)	School buildings	Indirect TEM, all lengths	222–1599 s/cm ³		
Guillemin et al. (1989)	Nonschool buildings	Indirect TEM, all lengths	15.6–54.4 s/cm ³		
Sebastien et al. (1982)	Paris, France	Indirect TEM	0.2–170 ng/m ³	Damaged floor tile	
Reported in Nicholson (1989)	U.S. buildings 1974	Indirect TEM	Mean 48 ng/m ³	Sprayed on friable and cementitious	
Reported in Nicholson (1989)	Paris, France, 1976–1977	Indirect TEM Indirect TEM	Mean 217 ng/m ³ Mean 35 ng/m ³	Cementitious	
Reported in Nicholson (1989)	U.S. schools 1980–1981	Indirect TEM	Mean 183 ng/m ³	Chrysotile	
Reported in Nicholson (1989)	U.S. schools 1980–1981	Indirect TEM	Mean 61 ng/m ³	71% chrysotile; 29% amphibole	
Reported in Nicholson (1989)	Ontario, Canada, 1982	Indirect TEM	Mean 2.1 ng/m ³	98% chrysotile 2% amphibole	
Reported in Nicholson (1989)	Ontario, Canada, 1977–1982	Indirect TEM	Mean 1.1 ng/m ³	87% chrysotile 13% amphibole	
Reported in Nicholson (1989)	UK buildings 1983–1985	Indirect TEM	Mean 1.5 ng/m ³	Surfacing	
				Surfacing	
				Surfacing	

TABLE 8. Fiber Concentrations in Residential and Source-Oriented Areas (Gaultier et al., 2009)

Category of fibers	Fiber length, μm	Fiber width, μm	Aspect ratio	Composition	Minimum count, f/cc	Maximum count, f/cc
All fibers	All	Any	$\geq 3:1$	Any	0.0000063	0.0000292
Regulated fibers	>5	<3	$\geq 3:1$	Any	0.0000058	0.0000284
Asbestos fibers	Any	Any	$\geq 3:1$	Asbestos	0.0000010	0.0000125
Regulated asbestos fibers	>5	<3	$\geq 3:1$	Asbestos	0.0000010	0.0000125
Fibers other than asbestos	Any	Any	$\geq 3:1$	Nonasbestos	0.0000016	0.0000218
Fibers other than asbestos	>5	<3	$\geq 3:1$	Nonasbestos	0.0000013	0.0000218

on analog geologic maps, that total igneous + metamorphic rocks worldwide are roughly one-fourth volcanic, one-fourth intrusive igneous, and one-half metamorphic, by area. Assuming amphiboles are predominantly limited to metamorphic and nonvolcanic igneous rocks, and if on the order of half of these rocks are amphibole-bearing (e.g., for the I-type granites of the Lachlan Fold belt of southeast Australia, Chappell and White [1992] estimated that approximately half are metaluminous and contain the amphibole hornblende), then approximately $0.5 \times 0.75 \times 17\% = 6\%$, by area, of exposed rocks in the conterminous United States are amphibole-bearing. This value is a little more than half that estimated by Thompson et al. (2001), but this estimate and the estimate of Thompson et al (2001) are within a half an order of magnitude and, given the uncertainties, should not be considered to be different from one another.

Regarding the proportion of amphibole relative to other minerals in rocks of the earth's crust, Klein (1993) estimated, based on models of the composition of the crust, that amphiboles constitute 5% of Earth's bulk (continental plus oceanic) crust overall. Similarly, based on the chemical compositions of igneous and metamorphic rocks exposed on the continents, Nesbitt and Young (1984) estimated that, where present, these surficial rocks comprise approximately 2% amphibole by volume. If one assumes that amphibole constitutes 2% of the 6% of the area of the conterminous United States underlain by rocks that are amphibole-bearing, there would be approximately 10,000 km² of amphibole in the United States subject to erosion. In the conterminous

United States, average denudation rates are close to 21 m/million years (Wilkinson and McElroy, 2007), yielding an annual release of more than 400,000 tons of amphibole; this yields, by erosion, 51 kg amphiboles per square kilometer per year for the conterminous United States. In mountainous regions where amphibole and serpentine may occur, the erosion rates may be higher such that these estimates may be considered a minimum. For example, in the Sierra Nevada, rates as high as 500 m/million years have been measured (Granger and Schaller, 2014).

It is possible to make an independent, order-of-magnitude estimate of the land area in the conterminous United States that is underlain by amphibole asbestos-bearing rocks, if one again assumes that amphiboles are dominantly restricted to metamorphic and nonvolcanic igneous rocks. The land areas of these occurrences likely span several orders of magnitude. To make an order-of-magnitude estimate (i.e., plus or minus unity on a log₁₀ scale) of the characteristic area of an asbestos occurrence, one first needs to consider reasonable bounds. Although no estimate has been made, our field experience suggests that the characteristic area is likely larger than 0.1 km² (an area ~ 300 m on a side), but is likely less than 10 km², suggesting the characteristic area of a given occurrence is on the order of 1 km². A rough order-of-magnitude estimate of the total area of the deposits of asbestos deposits in the conterminous United States is then 860 km²; letting on the order of half of these be amphibole (as opposed to serpentine) asbestos occurrences, then the corresponding area of amphibole asbestos occurrences is 430 km². Chrysotile

TABLE 9. Representative Concentrations of Airborne EMPs in Urban Areas

Reference	Location	Protocol	Concentration	Likely source	Probable EMP identity
Gualtieri et al. (2009)	Italian cities	NIOSH 7402 PCME	0.000006–0.00003 f/cm ³	Unknown	Chrysotile
Kovalevskiy and Tossavainen (2006)	Moscow, Russia	NIOSH 7402 PCME	BD (<0.001) to 0.002 f/cm ³	Unknown	Chrysotile
Lim et al. (2004)	Seoul and Incheon, Korea	NIOSH 7402 PCME, l > 0.2 μm	0.0052–0.0074 f/cm ³	Unknown	Chrysotile
Sebastien et al. (1986)	Montreal and St Etienne, Canada	NIOSH 7402, all widths	0.0009 f/cm ³	Unknown	Asbestos
Marfels et al. (1987)	German cities	SEM l > 5 μm, all widths	0.0001–0.002 f/cm ³	Industry, brakes, ACM	Asbestos
Marfels et al. (1987)	German cities	SEM l > 5 μm, all widths	0.001–0.01 f/cm ³	Unknown	Not asbestos
CPSC (1987)	San Francisco, CA	ISO 10312	0.005 s/cm ³	Unknown	
CPSC (1987)	San Francisco, CA	ISO 10312, l > 5 μm	BD	Unknown	
CPSC (1987)	Cleveland, OH	ISO 10312	0.006 s/cm ³	Unknown	
CPSC (1987)	Cleveland, OH	ISO 10312, l > 5 μm	BD	Unknown	
CPSC (1987)	Philadelphia, PA	ISO 10312	0.004 s/cm ³	Unknown	
CPSC (1987)	Philadelphia, PA	ISO 01312, l > 5 μm	BD	Unknown	
Reported in Nicholson (1989)	U.S. cities 1969–1974	Indirect TEM	Mean 3.3–13 ng/m ³	Chrysotile	
Reported in Nicholson (1989)	Paris, France, 1974–1975	Indirect TEM	Mean 0.96 ng/m ³	Chrysotile	
Reported in Nicholson (1989)	U.S. cities 1980–1981	Indirect TEM	Mean 6.1 ng/m ³	Unknown	
Reported in Nicholson (1989)	Toronto, Canada, 1980–1981	Indirect TEM	Mean 0.83 ng/m ³	Unknown	
Reported in Nicholson (1989)	Southern Ontario, Canada, 1980–1981	Indirect TEM	Mean 0.20 ng/m ³	Unknown	
Reported in Nicholson (1989)	German cities, 1982	ISO 10312	Mean 2.6 f/cm ³	Amphibole	
Reported in Nicholson (1989)	German cities, 1982	ISO 10312	Mean 2.8f/cm ³	Chrysotile	
Health Effects Institute (1991)	Outside schools, 1986–1988	NIOSH 7402 PCME	Median 0.00005–0.0003 f/cm ³	Unknown	
Health Effects Institute (1991)	Canadian cities	NIOSH 7402 PCME	Median 0.0001–0.0007 f/cm ³	Unknown	
Health Effects Institute (1991)	Urban U.S. school sites	Direct TEM	0–3.9 ng/m ³	Unknown	
Holt and Young (1973)	London, Reading, Rochdale, UK	TEM	Reported only as present	Unknown	
Holt and Young (1973)	Bochum, Dusseldorf, Germany; Prague, Czech Republic	TEM	Reported only as present	Unknown	
Holt and Young (1973)	Pilsen, Johannesburg, South Africa	TEM	Reported only as present	Unknown	
Holt and Young (1973)	Reykjavik, Iceland	Indirect TEM	0.1–10 ng/m ³	Unknown	
Nicholson et al. (1975)	50 U.S. cities	Indirect TEM	0.1–100 ng/m ³	Unknown	

TABLE 10. Representative Background Concentrations of Airborne EMPs: Rural Areas

Reference	Location	Protocol	Concentration	Likely source	Probable EMP identity
Cualtieri et al. (2009)	Italy	NIOSH 7402 PCME	BD (< 0.000001 f/cm ³)	Unknown	Unknown
ATSDR (2011)	El Dorado Hills, CA	NIOSH 7402 PCME	0.00001–0.0001 s/cm ³	Unknown	Unknown
Ladd (2005)	El Dorado Hills, CA (area 1)	NIOSH 7402 PCME	0.000290–0.00197 s/cm ³	Soils	Amphibole
Ladd (2005)	El Dorado Hills, CA (area 2)	NIOSH 7402 PCME	0.00285–0.00440 s/cm ³	Soils	Amphibole
Ladd (2005)	El Dorado Hills, CA (area 3)	NIOSH 7402 PCME	0.000284–0.00177 s/cm ³	Soils	Amphibole
Ladd (2005)	El Dorado Hills, CA (area 1)	I > 0.5 µm, l/w > 3 (AHERA)	0. 000570–0.00289 s/cm ³	Soils	Amphibole
Ladd (2005)	El Dorado Hills, CA (area 2)	I > 0.5 µm, l/w > 3 (AHERA)	0.000856–0.00586 s/cm ³	Soils	Amphibole
Ladd (2005)	El Dorado Hills, CA (area 3)	I > 0.5 µm, l/w > 3 (AHERA)	<0.000880–0.00624 s/cm ³	Soils	Amphibole
Lim et al. (2004)	Korea	NIOSH 7402 PCME	0.0003 f/cm ³	Unknown	Unknown
ATSDR (2009b)	Sapphire mine, NC	ISO 10312 PCME	BD (< 0.0001 s/cm ³)	Unknown	Unknown
Reported in Nicholson (1989)	Rural Switzerland	Indirect TEM	mean 0.74 ng/m ³	Unknown	
Reported in Nicholson (1989)	Rural Austria	Indirect TEM	BD		
Kohyama (1989)	Agricultural Japan	Indirect TEM	0.08–0.29 ng/m ³		

TABLE 11. Representative Concentrations of EMPs in Surface Water and Groundwater

Reference	Location	Protocol	Concentration	Likely source	Probable EMP identity
Hallenbeck et al. (1978)	Northern Illinois groundwater	Unspecified > 0.1 μm	0.04–0.32 MF/L	Unknown	Chrysotile
Hallenbeck et al. (1978)	Lake Michigan	Unspecified > 0.1 μm	0.029–0.67 MF/L	Unknown	Chrysotile
Hallenbeck et al. (1978)	Northern Illinois groundwater	Unspecified > 0.1 μm	0.08–0.48 MF/L	Background; AC pipe	Chrysotile
Hallenbeck et al. (1978)	Lake Michigan	Unspecified > 0.1 μm	0.058–0.55 MF/L	BACKGROUND; AC pipe	Chrysotile
Kay (1974)	Water for 22 municipalities, Ontario, Canada	Unspecified	0.0136 – 3.87 MF/L	unknown	Amphibole
Kay (1974)	Lake Superior, Duluth, MN	Unspecified	1–100 MF/L	Mine tailings in Lake Superior	Chrysotile
Oliver and Murr (1977)	New Mexico Rio Grande Valley	Unspecified	BD–2190 MF/L	Naturally occurring asbestos	Amphibole
Oliver and Murr (1977)	33 Municipal water supplies	Unspecified	BD–1680 MF/L	Naturally occurring asbestos	Chrysotile
Oliver and Murr (1977)	After cement pipe	Unspecified	and AC pipe	AC pipe	Chrysotile
Cook et al. (1975)	Duluth Lake Superior	ar \geq 3	15–644 MF/L	Mine tailings in Lake Superior	Amphibole
McMillan et al. (1977)	Southern Lake Michigan	Unspecified	0.5–4.6 MF/L	Unknown	80% Chrysotile
Webber et al. (1989)	Woodstock, NY	Unspecified	0.725–1850 MF/L	Detonated AC pipe	Chrysotile and crocidolite
Wei et al. (2013)	Rural China wells	ISO 14966	0.94–50.2 MF/L	Naturally occurring riebeckite	Riebeckite
Wei et al. (2013)	Rural China surface water	ISO 14966	15.1–399 MF/L	Naturally occurring riebeckite	Riebeckite

occurrences would then occupy a similar total area. From these estimates, it is possible to calculate the proportion of land area of the United States underlain by amphibole asbestos-bearing rock, relative to the total amphibole-bearing rock calculated previously, as $430 \text{ km}^2 / (0.06 \times 7.78 \times 10^6 \text{ km}^2)$ which is approximately 0.001, or 0.1%. In other words, on average, one might expect to find amphibole asbestos in 1 out every 1000 occurrences of amphibole minerals. Since not all asbestos occurrences are known, this estimate is a minimum.

Given that the ratio of finding amphibole asbestos-bearing rock relative to amphibole-bearing rock is on the order of 1/1000, and given that 400,000 tons of amphibole is released by weathering each year, then on the order of 400 tons of amphibole asbestos is released by weathering annually. Normalized to the area of the conterminous United States, approximately 50 g amphibole asbestos would be obtained from the erosion of 1 km^2 per year. Finally, if the area proportion of total asbestos to total land area is $860 \text{ km}^2 / (7.78 \times 10^6 \text{ km}^2)$, then the likelihood of encountering rocks, in the conterminous United States, that contain asbestos is on the order of 0.0001 or 0.01%.

Hendrickx (2009) summarizes probable asbestos locations in the four easternmost states in Australia. By assuming a denudation rate of 5 m/million years and a surface area of 5803 km^2 of ultramafic rock (0.2% of the land area) containing 0.1% asbestos, Hendrickx (2009) estimated that 72.5 tons of asbestos is released from this region annually into the air and water through erosion. If one included all amphibole and serpentine, these numbers might have been larger. Normalizing this quantity of asbestos to the land area considered by Hendrickx (2009) yielded 24 g asbestos per square kilometer per year. Despite the differences in denudation rates, the Hendrickx (2009) estimate for erosion of asbestos from eastern Australia (24 g asbestos/ km^2) and our estimate for the conterminous United States (50 g/ km^2) are within half an order of magnitude. Given the uncertainties associated with these estimates, which rely on

many assumptions, these two estimates should not be viewed as different.

Hiller (2001) reported amphibole in suspended sediment in a river on Scotland's east coast, confirming that where amphiboles and serpentine occur in soils, they may also be found in waterborne sediment. These would be most abundant in mountainous areas where erosion rates are high, and in arid regions where chemical weathering is low. In humid climates where chemical decomposition is active, amphibole and serpentine minerals are more likely to be altered to clays, amorphous silica, and other minerals, and are therefore likely to be less abundant in suspended sediment than they are in the underlying rock, but are not absent. In Fairfax County, Virginia, analysis of undisturbed forest soil overlying unexposed mafic rock found amphibole in all soil horizons (Martin Rabenhorst, personal communication); similar results characterize soils developed over mafic rocks in Middletown Valley, Maryland (Coffman, 1972). It should be noted, however, that there is little information on the size and shape of amphiboles found in soils in humid climates.

The occurrence of serpentine is more restricted than that of amphibole. Its two primary occurrences in the United States are in discrete discontinuous bodies found throughout the Appalachians and Rocky Mountains, in the alteration of many iron- and magnesium-rich rocks commonly referred to as ultramafic rocks, and in some metamorphosed limestone. The abundance of ultramafic rock in the conterminous United States has been estimated by Peucker-Ehrenbrink and Miller (2002) to be approximately 0.0015. Chrysotile may be found in many of these locations but in small amounts.

The entrainment of asbestos into the air from soil and weathered rock, in the absence of anthropogenic processes, is primarily a function of wind speed and turbulent intensity (the ratio of the standard deviation to the mean of the wind speed) in the boundary layer between the earth and the atmosphere (Lee et al., 2011). Data also showed that the rate

at which asbestos particles are entrained in turbulent air decreases with increasing moisture content of the soil. Once entrained, particles may be transported substantial distances by atmospheric dispersion plumes.

The effectiveness of the natural dispersal of asbestos is demonstrated by analyses of ice in Greenland and Antarctica, where ice preserves atmospheric particles from times before the widespread use of asbestos. Kohyama (1989) noted that chrysotile was identified in ice from Antarctica that is 10,000 years old. Gaudichet et al. (1986) also detected amphibole from Antarctica Dome C, and Bowes et al. (1977) reported asbestos from the Greenland Ice cap.

Although rapid mass movements such as landslides and other nonanthropogenic erosional processes contribute amphibole and serpentine EMP to air and water, as the data from the ice caps demonstrated, human disturbance of rock and soils containing amphibole and serpentine is probably also an important contributor of EMP to ambient air and water. This is described in the following sections.

Release of asbestos and cleavage fragment EMPs from mining and excavation of rock
Excavation of rock takes place during construction of building foundations, roads, and below-ground structures such as tunnels and mines. In addition, rock is mined, crushed, and used as aggregate for riprap, roads, railroad beds, concrete, and road metal, and macadam road surfaces. In 2012 alone, 1.24 billion metric tons of crushed stone was mined in the United States for these purposes (Willett, 2012).

In the mining and crushing process and application of crushed stone, EMP may be released into the air. In addition to crushed stone, amphibole and serpentine may be found in other industrial minerals and mineral products, for example, industrial talc, vermiculite, play sand, and calcium carbonate, and distributed into the ambient environment when their use creates dust. They are also known to occur as gangue (waste) material in some vermiculite, gold, copper, cosmetic talc, olivine, wollastonite, and iron ore deposits, where they are separated at the mine or mill and normally

disposed of nearby. These may also be dispersed in air and water.

Representative data for ambient contributions from mining, excavation, and use of mined material are provided in Table 4. Numerous studies have been conducted on the levels of EMP in and around mining communities where asbestos is a known component of the rock, but few are available where asbestos is absent or rare. Two large mines where massive amphibole is abundant have been the subject of study: the Peter Mitchell Pit in Minnesota and the now-closed Homestake Gold Mine in North Dakota. At both locations, airborne and waterborne ambient EMP contributions arise from fragmented amphibole, not asbestos. In both cases, extensive grinding of the ore took place in the mill, producing large quantities of fragmented amphibole EMP in tailings. In Minnesota, these tailings were initially pumped into Lake Superior until tailing disposal was moved to the land. An air monitoring program in Silver Bay, where ore from the Peter Mitchell Pit was processed, found levels of 0.0048 f/cm^3 in 1975 and 0.00014 f/cm^3 in 1998. The EMP have been identified as fragments of amphibole cleavage fragments, as well as commercial asbestos from sources other than the mine (Wilson et al., 2008); Hwang et al. (2014) found that the dimensions of airborne EMP from the Peter Mitchell Pit were between 1 and $3 \mu\text{m}$ in length and 0.2 – $0.5 \mu\text{m}$ in width, dimensions consistent with cleavage fragments of amphibole. The concentration of airborne cleavage fragment EMP from mining activity reported in Table 4A is generally lower than from mining in asbestos-containing rock (Table 4B).

Elsewhere, occupational monitoring at mines, where rock containing abundant massive amphibole is disturbed, clearly demonstrated that amphibole cleavage fragment EMP are released into the air, although little is known about the extent of their entrainment into the ambient environment. Bailey (2004) found that exposures to EMP in granite quarries in which no asbestos occurred are readily quantified and not uncommon. Wylie et al. (1983) described the size and shape of airborne

amphibole cleavage fragment EMP from exposures to "trap rock" mined in Virginia, gold mining in North Dakota, and the Peter Mitchell Pit in Minnesota, all locations where asbestos is rare or unreported. Amphibole cleavage fragments and asbestos fibers may also be released during mining of marble (Berman, 2003).

Many serpentinites, when crushed, release some level of chrysotile in addition to fragmented EMP (Table 4B). In one study (Rohl et al., 1977), the use of serpentinite crushed stone for road aggregate resulted in the high ambient concentrations of EMP. Similarly, Cattaneo et al. (2012) noted that airborne chrysotile was detected emanating from serpentine quarries and processing facilities in Italy. Sakai et al. (2001) reported elevated levels of EMP associated with serpentinite stone quarries in Japan, where exposures as high as 384 f/cm³ were noted for EMP longer than 0.2 µm.

Libby, Montana, was home to the largest vermiculite mine in the world, a mine that operated for much of the 20th century. The ore itself contained on average about 5% amphibole gangue, primarily winchite, richterite, and tremolite, both asbestiform and nonasbestiform (Meeker et al., 2003). The gangue was separated from vermiculite and discarded. Gangue accumulated in waste piles, was used for road surfacing, and in other ways was widely distributed in a relatively dry region where dusty conditions are common. However, some amphibole remained in the mine concentrate that was sent to vermiculite expansion facilities, where more, but not all, amphibole was removed. The amphibole EMP-containing vermiculite has been widely distributed throughout the United States, primarily as insulation. Disturbance of amphibole-rich material in and around the town of Libby results in airborne concentrations that range from below detection (bd) to 1.72 f/cm³ (Adgate et al., 2011; Januch and McDermott, 2004). The airborne particle population included both amphibole asbestos and cleavage fragments in comparable proportions.

The release of asbestos into the ambient air was documented from asbestos mine sites in South Africa, Australia, China, Finland, Quebec,

and elsewhere. Representative data are provided in Table 4B. Chrysotile asbestos concentrations in the ambient air in and around the Quebec chrysotile mines, and as much as 6 km from the mine source, were extensively documented. Sébastien et al. (1984) indicated that ambient air concentrations were significantly elevated over typical urban levels. Anastasiadou and Gidarkos (2006) reported concentrations as measured by phase-contrast microscopy of 0.08–0.18 f/cm³ at distances up to 40 km from an asbestos mine; these levels were similar to concentrations in the mine area. Murai et al. (1997) studied the lungs of residents from a region of Japan where formerly active anthophyllite and chrysotile asbestos mines were present, and identified four amphiboles; two of four amphibole EMP detected were not associated with anthophyllite and chrysotile, so they probably originated from exposure to commercial material containing asbestos (amosite and crocidolite). Luo et al. (2003) found environmental exposure in China, near crocidolite mines, from blue clays used for stucco and paint and from the production of clay ovens from friable crocidolite-bearing rock. Although no ambient airborne concentration levels are provided, Luo et al. (2003) demonstrated that the area is dusty and occupational exposure in the mine was as high as 25 f/cm³. Exposure extended from the source into the ambient environment over 200 km² in which 68,000 individuals reside.

Data in Table 4B make it clear that the mining of asbestos-containing rock may result in high airborne concentrations in the area surrounding the mine, including, in one case, up to 40 km away. However, the area affected depends on many factors, such as climate, wind levels, and precipitation, and therefore varies substantially from location to location as well as with time.

Ambient amphibole and serpentine particles derived from soil disturbance Amphibole and serpentine EMP in soils may readily be released into ambient air by any human activity that disturbs soil and creates dust, such as agriculture, driving vehicles on unpaved roads, and construction. Tegen et al. (1996)

reported that 50% of total atmospheric dust mass globally originates from soils disturbed by human activity. Green et al. (1990) found amphibole in airborne mineral dusts associated with farming in Alberta, Canada, and Voisin et al. (1994) detected ambient EMP tremolite in Afghanistan. Kalderon-Asael et al. (2009) reported amphibole and serpentine in atmospheric particles suspended over Israel, and attributed its source to soils from the arid and semi-arid dust belt that extends from 20 to 30° N latitude around the world. A type of soil "mining" occurs in and around this dust belt where soil is excavated for a variety of household uses and is disturbed during agricultural activities, and a number of air monitoring studies are available from these locations (Table 5).

In Turkey, Greece, Corsica, Cyprus, and New Caledonia, white soils contain tremolite asbestos. Their use as whitewash is associated with elevated occurrences of asbestos-related diseases (Constantopoulos, 2008; Metintas et al., 1999, 1999b, 2002; Zeren et al., 2000; Langer et al., 1987; Sakellariou et al., 1996; Senyigit et al., 2000). In New Caledonia, both tremolite-asbestos and chrysotile were found in the soils and in local serpentinite quarries (Luce et al., 2000; Baumann et al., 2007, 2011). In Afghanistan, no asbestos-related diseases were reported, but tremolite EMP detected in soil dust and pleural plaques may be associated with exposure to it (Voisin et al., 1994). Measurements of outdoor EMP concentrations in Turkey were reported by Metintas et al. (2002) at levels of 0.009–0.040 f/cm³ in villages near Eskisehir where soil mining provided white wash. Massaro et al. (2012) reported airborne tremolite EMP in the area of Basilicata, Italy, at levels from <1 to >200 f/cm³ where soils overlying serpentinite are disturbed. Constantopoulos (2008) demonstrated residential exposure to EMP in Metsovo, Greece, of >200 f/cm³ in the preparation of white wash and 1–4 f/cm³ while cleaning floors inside houses that used it on the walls. These levels are probably typical of such exposures, which are similar to mining and excavation of rock containing asbestos (Table 4B).

Throughout the western United States, arid regions predominate from the Great Plains to the west coast. In this area, soil disturbances result in atmospheric entrainment of dust. In two arid areas of California, disturbance of soil resulted in significant release of asbestos and amphibole cleavage fragment EMP (Table 5). Addison et al. (1988) demonstrated that a mass concentration of asbestos in dry soils at low as 0.001% was capable of producing in excess of 0.1 f/cm³ under fairly routine conditions of disturbance. In the Clear Creek Management Area of California, chrysotile is readily released into the atmosphere when soils overlying the chrysotile-rich New Idria serpentinite are disturbed, yielding airborne concentrations that are similar to occupational exposures (U.S. EPA, 2008b; Cooper et al., 1979). The soils in and around El Dorado Hills, California, an arid area, contain fragmented amphibole EMP (tremolite-actinolite), and tremolite asbestos was also detected from the area. Soil disturbance during construction of houses, schools, and roads released amphibole EMP-bearing dusts into the ambient air of this community (Ladd, 2005, 2011a) at levels similar to those at Silver Bay where nonasbestos EMP are also found (Table 3A). Buck et al. (2013) reported naturally occurring asbestos actinolite and magnesiohornblende in soils from southern Nevada that are easily disturbed and entrained into the air by recreational activities.

Ambient Asbestos Derived from Asbestos Use

According to the U.S. Geological Survey (USGS), between 1900 and 2012, more than 31 million metric tons of asbestos (all types) was consumed in the United States, either from domestically mined asbestos (which ceased in 2002) or from imports, and incorporated into products (Virta, 2005). Virta (2006) estimated world asbestos production and consumption between 1900 and 2003 to be approximately 181 million metric tons. The USGS asbestos end-use data, covering the period 1965 to 2003, indicated that although in some uses

asbestos was consumed and discarded, such as in friction products or packing and gaskets, 60% of the asbestos was incorporated into durable products such as asbestos cement pipe, asbestos cement sheet, flooring products, roofing materials, and insulation that remain in place for long periods of time (Virta, 2006). Although no apparent estimates have been made on how much asbestos remains in buildings, this material, when disturbed, continues to contribute to ambient asbestos until it is removed and buried in landfills.

Industrial sources that use or have used asbestos to fabricate asbestos-containing materials contribute significant numbers of fibers to the ambient environment (Table 6). Awad (2011) reported EMP concentrations as high as 5.4 f/cm³ at locations 0.5 to 7 km away from asbestos concrete manufacturing plants in Egypt. Data suggest that high PCM-based concentrations of EMP exist close to both operating and closed asbestos concrete facilities and waste disposal areas. Maule et al. (2007) summarized the observed EMP concentrations in Casale Monferro, Italy, with a large asbestos concrete pipe plant. Data indicated that the asbestos PCME concentrations were as high as 0.011 f/cm³ within 400 m of the plant and 0.001 f/cm³ in a part of the city remote from the plant. Magnani et al. (1998) found higher-than-expected concentrations of asbestos bodies in lung tissue from residents of Casale who had no occupational exposure. At the time of the measurements, the production levels were low.

On average, 13 million lb of friable asbestos was reported to the U.S. EPA as disposed of, or otherwise released, annually between 2003 and 2012 (U.S. EPA Toxic Release Inventory, 2012). Sources of this material include building materials, but this material may also derive from removal of legacy occurrences such as waste dumps, transport sites, road and yard surfaces, and so on. In Jefferson Parish, Louisiana, the U.S. EPA remediated more than 1400 properties where crocidolite-bearing scrap was dispersed (Case and Abraham, 2009).

There are no apparent data available on the amount of asbestos that remains today in durable products in the United States. Some has been discarded during renovation and replacement. If only asbestos from nonfriable materials remains today (and all other asbestos-containing materials are in landfills), and if half of the asbestos initially incorporated into the durable products has been removed and buried in landfills, then an estimated 10 million metric tons remains in the built environment in the United States alone. Whatever the amount is, asbestos in buildings continues to contribute to ambient asbestos levels when disturbed. Higher-than-anticipated concentrations in ambient air were found from asbestos-containing buildings damaged by the 2011 tsunami and earthquake in Japan (Japan Daily Press, 2013), from a fire in England (Bridgman, 2001), and from the collapse of the World Trade Center (Nolan et al., 2005). As late as 2011 the U.S. EPA was citing building demolition activities in Philadelphia for releasing asbestos into the air (U.S. EPA, 2011b).

The asbestos in friable construction material, if disturbed, may contribute to asbestos in indoor air as long as it remains in the building. Hundreds of measurements of asbestos in the indoor air in occupied buildings in which asbestos-containing materials are found have been made throughout the United States and the world (see Table 6 for representative studies). Nicholson (1989) summarized ambient air concentrations in buildings containing asbestos between 1974 and 1985, and the Health Effects Institute (1991) added comprehensive overview of the literature that included studies published up to 1991. Data on indoor air concentrations are often presented in units of nanograms per cubic meter (ng/m³) instead of f/cm³, and such units of measure are not directly comparable. However, data show that there are usually EMP in the air if asbestos is found in friable building materials, although the mean levels may vary over several orders of magnitude. Commins (1985) reviewed indoor air data and concluded that long-term overall indoor concentrations for

PCME EMP could be in the range of approximately 0.0002 to 0.001 f/cm³ with a typical figure of 0.0005 f/cm³. The Health Effects Institute (1991) data reported a range from 1599 to <0.0001 f/cm³ where detected; the larger concentrations were derived by using the Yamate et al. (1984) method, which does not have length restrictions. Sebastien et al. (1982) measured asbestos released from normal wear of asbestos-containing vinyl tiles, so even encapsulated nonfriable asbestos may be released into indoor air through normal wear, although at low levels. Nonetheless, if indoor air contains asbestos, some of it penetrates the outside ambient air, and friable asbestos remains in buildings today.

Chrysotile is the predominant form of asbestos present in, and released from, asbestos-containing piping, but crocidolite is also found. Webber and Covey (1991) reported that there were, at the time of writing, more than 2×10^6 miles of asbestos-containing piping worldwide and approximately 400,000 miles of pipe carried U.S. public water. Asbestos contamination was found in the Village of Woodstock, NY, public water supply in 1985 at levels between 0.725 and 1850 MF/L; chrysotile constituted 90% of the measured asbestos fibers, and crocidolite 10% (Weber et al., 1989) (Table 11). The contamination was discovered during routine flushing of the water mains and was postulated to be due to extensive corrosion of the pipe by water. Millette et al. (1979) concluded that asbestos is contributed everywhere there is asbestos-containing (AC) pipe but that the levels are likely to be low except under certain situations (such as Woodstock, NY) where water chemistry results in active decomposition of the pipe. Millette et al. (1979) estimated that in 1979 such conditions occur in about 16% of U.S. water utilities. Under these conditions concentrations as high as 10 MF/L were detected.

Urban air It is widely assumed that the ambient EMP found in urban air originated from use of building materials that contain asbestos, as well as from emissions from brake linings. However, this may not be the case. Gibbs and Pooley (2008) summarized data from

six studies on lung burden, from more than 200 individuals, and concluded that although chrysotile and amphibole EMP are present in almost all lungs and are higher in those living in urban areas than in rural areas, lung burdens include four- to fivefold greater nonasbestos fibers than asbestos. The studies of indoor air in buildings with asbestos-containing materials clearly demonstrated the presence of asbestos, and, in general, the EMP levels are higher than in urban outdoor air (Tables 6 and 7). Enterline (1983) estimated that the average level in the air in the United States as a whole is about 0.0015 ng/m³; for urban areas the average is 0.003 ng/m³, and for rural areas, the average is 0.0001 ng/m³. Although it is reasonable to assume that, given the extensive use of asbestos, some of the EMP in urban air arises from use of asbestos in building products, NOA and other EMP of amphibole and serpentine may also contribute to airborne EMP, especially in arid and semi-arid regions.

Van Orden et al. (1995), using analytical TEM, evaluated air samples from inside and outside buildings containing asbestos that were affected by the Loma Prieta earthquake in California in 1989. Data showed a significant inside/outside difference in EMP concentrations but no marked difference with respect to levels of fibers exceeding 5 μm in length. This study indicated that ambient air quality may not be substantially affected by the presence of asbestos in buildings, even when the buildings are subject to the disturbance of an earthquake. On the other hand, when buildings are destroyed by earthquakes, the case may be different. Reportedly, elevated levels of EMP were found in Japan attributed to asbestos-containing materials in buildings destroyed by the 2011 earthquake (Japan Daily Press, 2013)

Gualtieri et al. (2009) measured ambient EMP concentrations at a variety of Italian residential and source areas. The sources consisted of areas with some asbestos concrete construction materials in buildings and a remote area with serpentinite outcrops. All monitoring sites were at least 10 m from the side of a road. They analyzed the fibers by x-ray powder diffraction and PLM followed by a combination of TEM

and SEM analyses. Results are shown in Table 9 and illustrate the complexity of airborne particles in urban atmospheres. These data illustrate that almost all EMP were longer than 5 μm and that, in all dimensional categories reported the concentration of EMP other than asbestos exceeded that of asbestos.

Despite some uncertainty regarding the source of urban EMP and their mineral identity, it was found in every urban area globally that has been studied (see Table 7). Given the decline in use of asbestos since 1975, one might expect to see this reflected in declining concentrations in urban air. However, although virtually no asbestos has been used in building materials in the United States and Europe for almost 40 years, normal wear and tear, as well as demolition of buildings and structure fires, continue to contribute asbestos fibers to urban ambient air.

It is difficult to draw any conclusions regarding changes in ambient levels in urban areas over time because (1) the methods that have been used vary substantially, (2) imprecision in measurements is high due to low concentration levels, and (3) there are few recent data. The study by Marfels et al. (1987) on German cities detected the presence of EMP in all samples but at levels less than 0.002 f/cm³. Marfels et al. (1987) reported that EMP less than 0.1 μm in diameter accounted for 98% of all sizes of EMP, suggesting that they were primarily asbestos. An earlier study reported by Nicholson (1989) found levels as high as 2.8 f/cm³ in German cities. However, the higher concentrations of the early study include all widths, whereas the later study restricted widths to greater than 0.2 μm , illustrating the impact of differing definitions of fiber.

Over the course of the 20th century, mineral dusts in the atmosphere increased, particularly in urban areas. Lung burden studies of urban dwellers provide compelling evidence that even at the low levels found in most urban air, EMP of a variety of types are inhaled, deposited and retained in the lungs of all urban dwellers (Paoletti et al., 1987; Pooley et al., 1970; Langer et al., 1971; Selikoff and Hammond, 1970; Friedrichs et al., 1992;

Yamada et al., 1997; Churg and Wiggs, 1986; Churg, 1982; Cralley et al., 1968; Dodson et al., 1988; Stettler et al., 1991; Bowes et al., 1977; Churg and Warnock, 1980; Enterline, 1983; Gibbs and Pooley, 2008).

Airborne EMP background Some investigators measured EMP concentrations in areas far removed from known urban, industrial, mining, building, and geological sources of asbestos (Table 10). Measurements in these areas provide a general indication of the background levels that result from the long-range transport of EMP from both anthropogenic and geologic sources. It has been possible to identify EMP in many remote areas in the world.

Lim et al. (2004) measured fiber concentrations in several remote areas of Korea. Using NIOSH 7402, they reported an EMP concentration of 0.0003 f/cm³. Chatfield (1983) provided background for southern Ontario as 0.2 ng/m³. Litistorf et al. (1985) estimated background in Switzerland to be 0.23 ng/m³. Spurney et al. (1976) noted asbestos in ambient air from rural Germany. The U.S. EPA Agency for Toxic Substances and Diseases Registry (ATSDR) report (ATSDR, 2011a) indicated that background EMP concentrations in areas remote from any known sources of asbestos in El Dorado Hills, California, ranged from 0.000003 to 0.0000003 f/cm³ (probably PCME EMPs). The background concentration, when conditions were dry, was estimated to be between 0.00008 and 0.0008 f/cm³. Kohyama (1989) reported finding chrysotile in a remote Pacific Island.

However, there are also studies that did not find EMP in rural areas. Gualtieri et al. (2009) measured background concentrations in three areas of Italy thought to be free of sources and deposits of asbestos, and did not report finding EMP at a detection level of 0.0001 f/cm³. Felbermayer (1983) detected no EMP in rural Australia, and a study in North Carolina in a rural area near the Sapphire Mine did not detect EMP (ATSDR, 2009b). However, given the ubiquity of airborne mineral EMP, they are likely to be present at some level everywhere under some conditions. In 1984 the National Research Council (1984) estimated

that a reasonable long-term outdoor median concentration of asbestos (EMP) was in the range of 0.00002 to 0.0075 f/cm³. Considering only EMP longer than 5 µm and visible by optical microscopy, the median concentration was estimated to be 0.00007 f/cm³.

Interpreting airborne concentration data
The use of differing criteria for designation of EMP as fibers, the use of different protocols to measure concentrations, and the variability in sampling schemes remain serious impediments to understanding ambient concentrations of asbestos and nonasbestos EMP. First, the methods do not distinguish asbestos fibers from cleavage fragments. Second, concentrations are indices of exposure and do not provide a complete characterization of airborne or waterborne concentrations. Third, because most cleavage fragment EMP have widths greater than 0.25 µm if they are 5 µm or longer, PCME data include them all. On the other hand, because most asbestos fibrils are less than 0.25 µm in width, PCME data exclude a large portion of them. For this reason, the index of exposure established by optical microscopy is not comparable between these two types of particles. Fourth, concentration data that include all lengths are not comparable to those that include only lengths longer than 5 µm or 0.5 µm for that matter, and fifth, data expressed as ng/m³ are not comparable to data expressed as f/cm³. Sixth, sampling protocols are highly variable and rarely consider temporal variability. Data provided by Lee and Van Order (2008, Table 2) are instructive in understanding the complexity of the fiber nomenclature, the incompatibility among fiber definitions, and the magnitude of the variances expected for low concentrations of airborne fiber. Taken together, these issues render the absolute values of low concentrations of ambient airborne EMP essentially meaningless and they are provided in Tables 3–10 only to demonstrate that they were detected. These issues need to be addressed in future studies before data on ambient airborne asbestos or other EMP concentrations can be used for predictive risk assessment or as baseline data to

evaluate changes in our ambient environment over time.

Atmospheric transport of EMP Once asbestos enters the air, its fiber bundle size distributions are not necessarily stable. Particles settle out by gravity or aided by precipitation. Due to the action of wind, clusters or bundles of fibers may breakdown into individual fibrils. Some clusters also settle due to their high aerodynamic diameters. Accordingly, the mix of EMP commonly observed in building remediation programs or disturbed natural deposits may not persist for substantial distances for the release points. As the distance from the emission point increases, the population of aerosol asbestos fibers gradually shifts toward higher proportions of individual fibrils of narrow width, making them more difficult to detect by optical microscopy.

The behavior of EMP in air is largely determined by their aerodynamic diameter. Burke and Esmen (1978) proposed an equation for estimating the aerodynamic diameters of EMP. Figure 1 illustrates the relationship between aspect ratio and aerodynamic diameters of EMP having physical diameters of 0.3, 1, and 3 µm (John Richards, personal communication). Their study predicts that for thin EMP, the aerodynamic diameter is effectively independent of length, such that extremely long narrow EMP may be transported as far as short EMP.

The transport distances of narrow EMP in ambient air have been shown to be large (U.S. EPA, 2002). Studies of PM_{2.5} particles (those with diameter of less than 2.5 µm) have clearly demonstrated transport distances of more than 500 miles due to the lack of any strong removal forces such as inertial impact, gravity settling, and/or electrostatic attraction. The physical forces are equally ineffective in removing atmospheric EMP having widths less than 0.5 µm, which includes most airborne asbestos fibers. Accordingly, remote occurrences of naturally occurring asbestos might influence wide areas and localized detection of EMP would not necessarily indicate a local source. Based on the aerodynamic characteristics of narrow EMP, they should be present in ambient air

in even highly remote areas such as Alaska and Greenland. Their identification in remote islands in the Pacific and Antarctic ice confirms this prediction (Bowes et al., 1977).

SOURCES AND MEASURED CONCENTRATIONS OF AMPHIBOLE AND SERPENTINE IN WATER

Asbestos and cleavage fragment EMP of amphibole and serpentine may enter surface water in the same manner they enter the air: erosion of natural materials, mining, and release from asbestos containing building materials. Representative data are presented in Table 11. Comprehensive reviews are provided by Millette et al. (1979) and Webber and Covey (1991). Asbestos EMP in drinking water have not been demonstrated to present a risk to human health (Truhaut and Chouroulinka, 1989; DHHS Committee, 1987; Gamble, 2008; Browne et al., 2005), but might enter the air through use of water for activities such as watering gardens, washing cars, or washing dishes that result in evaporation. Webber et al. (1988) measured airborne levels of chrysotile and amphibole in homes in Woodstock, NY, by using water that flowed through badly deteriorating asbestos cement pipes at greater than 2 f/cm^3 . Most of the fibers were attributed to the pipe, but the presence of EMP of the amphibole anthophyllite was attributed to nonanthropogenic sources. Thus, not only can waterborne EMP contribute to airborne EMP concentrations, but the converse may also be the case. Taken together, air and water widely distribute such particles.

The studies of the transport of amphibole cleavage fragments from iron mining operations in Minnesota are informative on the potential distribution by water of particulate generated by mining when tailings are disposed of in surface water. Fragmented amphibole EMP were identified in the Duluth, MN, municipal water supply at levels of 10 to 1000 MF/L (Cook et al., 1975). Flickinger and Standridge (1976) reported that Lake Superior water entering water treatment facilities in two

Wisconsin communities contain EMP in the range of 0.007–0.0003 MF/L for crystalline material identified by light microscopy, and 0.008–0.07 MF/L for silicate material identified by SEM. After water treatment, cleavage fragments are present but at reduced concentrations. Both of these studies were conducted during the time when more than 60,000 tons of taconite tailings containing abundant amphibole cleavage fragments were discharged into Lake Superior daily, a procedure now discontinued. It should be pointed out again here that these “fibers” are not asbestos fibers but amphibole cleavage fragments that meet the given definition of fiber.

Although amphibole and serpentine EMP are entrained into air by natural erosional processes, concentrations in surface water and groundwater from these processes are poorly known. However, there are a few studies that indicate the levels to be expected. A study of EMP in surface water in the Rio Grande Valley that supplies drinking water to New Mexico identified chrysotile at levels from <1 to >2000 MF/L attributable to erosion of serpentinite in the watershed (Oliver and Murr, 1977). Webber and Covey (1991) noted that asbestos in most natural streams and lakes is at less than 1 MF/L. However, concentrations of chrysotile ranging from 10 to more than 1000 MF/L in surface waters were measured in parts of the eastern United States, California, and Quebec (Canada), attributed in part to serpentinite bedrock. However, contributions to surface water from airborne particles cannot be ruled out as they are known to nucleate raindrops. EMP concentrations in the range of 15 to 86 MF/L were detected in rivers in New York and New Jersey, also likely from multiple sources. McMillian et al. (1977) reported chrysotile in Lake Michigan water that ranges over the course of a year from about 0.5 to 4.5 MF/L. Kay (1974) found asbestos in all surface water that supplies municipal water to 22 Ontario cities at concentrations that ranged from 0.136 to 1.9 MF/L, mostly chrysotile from sources thought to include both commercial and NOA. Hallenbeck et al. (1978) identified chrysotile in groundwater systems in northeast

Illinois, at levels between 0.04 to 0.32 MF/L, although serpentinite is not known in this part of the United States. Wei et al. (2013) found crocidolite from natural occurrences in drinking water from rural China.

A World Health Organization (WHO, 2003) report indicated that 25% of public water supplies in Canada have levels exceeding 1 MF/L for 25% of the population, levels exceeding 10 MF/L for 5% of the population, and levels exceeding 100 MF/L for 0.6% of the population. Median fiber lengths are in the range of 0.5 to 0.8 μm . In Quebec, mining of chrysotile and the abundance of serpentinite bedrock likely contributed to these levels, which are higher than found in the United States. WHO also reported that most drinking water supplies in the United Kingdom varied from nondetectable levels to up to 1 MF/L and that a few water supplies in the Netherlands noted concentrations up to 33 MF/L. The sources of the EMP are unknown.

Millette et al. (1979) provided hundreds of measurements of domestic water supplies from across the United States prior to entry into AC pipe. Excluding those likely to contain EMP cleavage fragments associated with disposal of tailings in Lake Superior, data demonstrated that while generally less than 1 MF/L, both amphibole and serpentine EMP are ubiquitous. A summary of available data provided by Webber and Covey (1991) affirms the ubiquity of these particles in surface water in major U.S. cities. Results indicated that prior to 1991 more than 80% of the major cities in the United States had EMP levels in drinking water that were less than 1 MF/L, 12% had levels between 1 and 10 MF/L, and the remaining 8% had levels exceeding 10 MF/L (method 100.1). The presently applicable maximum concentration limit for drinking water in the United States is 7 MF/L longer than 10 μm (U.S. EPA, 2014b). Although most U.S. cities have levels below the limit, the distribution by water of EMP of amphibole and serpentine, both natural and as commercial asbestos, is extensive.

In summary, water serves as means for dispersal of EMP. Use of surface water can result in the transfer of waterborne EMP to air. The

magnitude of airborne EMP that have been transported by water remains unknown.

TEMPORAL TRENDS

The USGS data indicate that consumption of asbestos in the United States has decreased from a high of 803,000 metric tons in 1973 to 1060 metric tons in 2012, and no asbestos has been mined in the United States since 2002 (Virta, 2004). Worldwide production has also decreased from a high of 4.8 million metric tons in 1977 (Virta, 2004). Building materials containing asbestos are aging, and it might be expected that as they age, their ability to encapsulate asbestos will decrease, making release of fiber into air and water more likely. Renovation of buildings and removal of asbestos-containing material also continue, inevitably contributing some amount of asbestos fiber to ambient air locally, as the recent U.S. EPA citation in Philadelphia and the experience in Japan attest. There is no reason to expect a decrease in release of EMP from mineral deposits in areas such as California and the Appalachian Mountains of the eastern United States. In fact, the disturbance of rocks and soils produced by expanding suburbs may expose fresh soil to erosion, as well as generating dust from human activity.

Although ambient airborne asbestos might be expected to decline over time as building materials containing asbestos are replaced, such a decline in ambient water concentrations is not likely in the near future. While production and installation of new asbestos-containing water pipes have ceased in the developed world, they continue in the developing economies. In much of the world, there remain large quantities of this piping in use. The reduction in the use of new asbestos-containing pipe is almost certainly being offset by the continuing deterioration both inside and outside of existing asbestos piping in some areas, especially in those areas with mildly corrosive pH levels. Other continuing sources of asbestos in ambient water include runoff from areas affected by improper land disposal of asbestos-bearing wastes and asbestos

fibers released from asbestos containing building materials. The possible continued release of asbestos fibers from wastes discharged into lakes is also a continuing source, as is contributions to surface water from EMP that provide nucleation for raindrops.

CONCLUSIONS

The global transport of low concentrations of EMP in air and water will continue even as the production of regulated asbestos is significantly reduced. Deterioration and demolition of asbestos-containing building materials and water pipes will continue to release asbestos. The expansion of the population into arid regions and the continuing human activities that disturb amphibole- and serpentine-bearing soils and rock will continue to create EMP-bearing dusts. Natural processes produce small particles of amphibole and serpentine. Taken together, these ensure that amphibole and serpentine EMPs will remain in the ambient environment well into the future. Because there is no standard definition for the term fiber, and because there is no recognized method for distinguishing asbestos from fragmented amphibole and serpentine in air and water, data derived from air and water analysis (1) are difficult to compare from study to study, (2) will record concentrations of both fragments and fibers, and (3) should not be assumed to measure asbestos exposure.

In interpreting these data, one needs to remember the words of Paracelsus: "Nothing is without poison" and "only the dose permits a poison not to be poisonous." Our extraordinary sensitive analytical techniques have enabled us to detect amphibole EMP and chrysotile in many varied environments. However, the levels may be very low, and to ascertain their potency as a "poison," risk assessments are necessary. However, there is no epidemiology on low-level exposures to asbestos such as those found in buildings, in schools, or in urban areas on which such assessment can be based. Further, given the wide variance in the airborne data both temporally and spatially, the

uncertainty in the identification of the particles being counted, and the wide variety of habits and sources, a meaningful assessment of risk from low level concentrations typical of ambient air remains elusive, particularly given the uncertainty associated with extrapolating a dose/response model derived from occupational exposure to a low-level exposure.

CONFLICT OF INTEREST

Author Wylie serves as a member of the Scientific Advisory Board for the National Stone Sand and Gravel Association. She has also served as a consultant in the mineralogy of industrial minerals to mineral producers and law firms. She has not served as an expert witness in litigation for more than 20 years. Author Candela has served as a consultant in the mineralogy of industrial minerals to mineral producers and law firms, and has served as an expert witness related to litigation.

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